

### **REMARKS**

The Official Action dated March 2, 2010 has been carefully considered. Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claim 5 is amended to recite the dehydration step and the subsequent polymerization steps are conducted in the same reaction vessel, as set forth in the specification at page 16, lines 24-26 and in the Examples. Claims 17 and 18 are amended to correct typographical errors. Claim 19 is presented to further define the dehydration step and support for claim 19 may be found in the specification, for example at page 17, lines 6-11. It is believed that these changes are fully supported by the original specification, whereby entry of the present Amendment is in order and is respectfully requested.

Claims 5, 7, 10-13, 17 and 18 were rejected under 35 U.S.C. 103(a) as being unpatentable over Miyahara et al, US 5,840,830. Although the Examiner asserted the claims were anticipated by Miyahara et al (page 3, line 3), it is believed that under 35 U.S.C. §103(a), the Examiner is asserting that Miyahara et al render the claims obvious. The Examiner asserted that Miyahara et al teach a process for producing a poly(arylene sulfide) (PAS) by polymerizing a sulfur source and a dihalo-aromatic compound in an organic amide solvent. The Examiner asserted that Miyahara et al teach many of the limitations of claim 5 and, while the Examiner acknowledged differences between the Miyahara et al process and the process of claim 5, the Examiner asserted that such differences did not prevent the finding of a prima facie case of obviousness.

This rejection is traversed and reconsideration is respectfully requested. Applicants submit that Miyahara et al do not teach or suggest, or make otherwise apparent to one of ordinary skill in the art, important limitations of the process of claim 5 which in combination provide PAS

with improved properties. That is, by carefully controlling the reactants and reaction conditions, the present process allows the production in a single reactor of a desirable and versatile PAS from relatively inexpensive reactants, providing an economically viable process. More particularly, the process for producing a PAS according to the present invention and as defined in claim 5 comprises four important process steps which are conducted in a single reactor and which in combination, provide a PAS having a desirable combination of improved properties as also set forth in claim 5.

Specifically, in the dehydration step (1), an organic amide solvent, and an alkali metal hydrosulfide and an alkali metal hydroxide, both as aqueous mixtures, are heated and reacted in a proportion of 0.95 to 1.02 mol of alkali metal hydroxide per mol of the alkali metal hydrosulfide, wherein distillate containing water is discharged to the exterior of the system and hydrogen sulfide formed upon the dehydration is discharged as a gas to the exterior of the system. As the discharge of hydrogen sulfide to the exterior of the system is directly linked with a weight loss of sulfur amount in the system, it is evident that the discharged hydrogen sulfide is not recycled into the reaction system.

In the subsequent charging step (2), the total number of mols of (i) alkali metal hydroxide formed with hydrogen sulfide upon the dehydration, (ii) the alkali metal hydroxide added prior to the dehydration, and (iii) the alkali metal hydroxide added after the dehydration is 1.015 to 1.050 mols per mol of the charged sulfur source and the mols of water are controlled to provide 0.5 to 2.0 mols of water per mol of the charged sulfur source. The amount of the charged sulfur source is calculated out in accordance with the equation:  $[\text{Charged sulfur source}] = [\text{Total moles of sulfur charged}] - [\text{Moles of sulfur volatilized out after dehydration}]$ , again evidencing that the discharged hydrogen sulfide is not recycled into the reaction system. Additionally, the

polymerization is conducted in the defined first-stage polymerization step (3) and the defined second-stage polymerization step (4) to provide a polymer as claimed, in the same reactor in which the dehydration step is conducted.

The combination of the defined steps (1)-(4) is important in providing a PAS polymer product in high yield and having good melt viscosity, low yellowness index, low dimer byproduct, good reactivity with aminosilane coupling agents, and low volatility from the alkali metal hydrosulfide and alkali metal hydroxide reactants in an economically viable process.

In this regard, the Examiner's attention is directed to the enclosed article from *Azo Materials* dated November 2005 and the enclosed *Business Wire* posting dated August 12, 2010, both of which are directed to a poly(phenylene sulfide) (PPS) plant of Fortron Industries LLC, a joint venture between the assignee of the present application, Kureha Corporation, and Ticona Engineering Polymers. The articles describe the Fortron PPS as a high performance polymer and indicate that the plant continues to increase production of PPS using "Kureha's advanced environmentally friendly 'one unit' process", the *Azo Materials* article further noting the process as making the plant highly efficient. These articles demonstrate that processes for PPS are technically important as is a process conducted in a single reactor, i.e., a one unit process, as recited in claim 5.

The present process differs from Miyahara et al in several respects. While each difference alone may appear to the Examiner as minor, these differences in combination are significant and, along with the other limitations of claim 5, provide a process and a high quality PPS product that are neither taught, suggested nor apparent from the teachings of Miyahara et al. It is preferred to use the claimed alkali metal hydrosulfide and an alkali metal hydroxide as starting reactants as they are less expensive than the traditionally employed alkali metal sulfide.

However, in the past, it has been difficult to produce a polymer having a good combination of properties in a stable manner using these reactants, as described in the present specification, beginning at page 2, line 17. Applicants discovered, as further described in the present specification, for example beginning at page 17, line 6, the alkali metal hydroxide unexpectedly reacts with the organic amide solvent in the dehydration heat treatment to form an alkali metal alkylaminoalkanoate, and the alkali metal hydrosulfide exists in the system in the form of a complex with the alkali metal alkylaminoalkanoate. While a part of the alkali metal hydrosulfide reacts with water to form hydrogen sulfide and an alkali metal hydroxide, the resulting hydrogen sulfide is discharged outside the system. The steps recited in claim 5 control these various reactions arising from the use of the alkali metal hydrosulfide and alkali metal hydroxide reactants to provide a stable reaction and a polymer product having a consistently good combination of properties as recited.

Miyahara et al disclose and exemplify a process for producing poly(arylene sulfide) wherein an alkali metal sulfide such as sodium sulfide is reacted with a dihaloaromatic compound, and wherein hydrogen sulfide vaporized off during the dehydration step is recovered and reused. Miyahara et al indicate that the sodium sulfide can be produced by in situ reaction of sodium hydrosulfide and sodium hydroxide (column 4, lines 61-64) and that these reactants can be used in equimolar amounts (column 6, lines 49-53). Importantly, Miyahara et al fail to exemplify any process wherein sodium hydroxide and sodium hydrosulfide are employed as reactants as required by present claim 5 or wherein sodium sulfide is formed in situ.

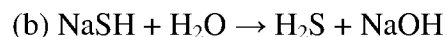
Importantly, if the process of Example 1a of Miyahara et al uses an alkali metal hydrosulfide and an alkali metal hydroxide in equimolar amounts in place of the exemplified sodium sulfide in the dehydration reaction, the proportion of alkali metal hydroxide to alkali

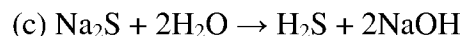
metal hydrosulfide is not in the ratio of 0.95 to 1.02 mol as required by claim 5. That is, Example 1a of Miyahara et al was conducted in the same manner as in Comparative Example 1a (column 12, lines 16-18) with respect to the amount of sodium sulfide, wherein “3,800 g of sodium sulfide pentahydrate containing 46.20 wt. % of sodium sulfide ( $\text{Na}_2\text{S}$ )” (column 10, lines 58-59). The number of moles of sodium sulfide is  $(3,800 \times 0.4620)/78.05 = 22.49$  moles, as the molecular weight of sodium sulfide is 78.05. Thus, the alkali metal hydrosulfide and the alkali metal hydroxide used in equimolar amounts, in place of the exemplified sodium sulfate, would be 22.49 mol of each. However, Example 1a of Miyahara et al *adds an additional 30 g of 97% NaOH in the dehydration step*, which comprises an additional 0.72 mol NaOH, resulting in a NaOH/NaSH ratio of  $(22.49 + 0.72)/22.49$ , which is equal to 1.032. This ratio is outside the NaOH/NaSH range of 0.95 to 1.02 required in the process of claim 5. Examples 1b, 2 and 5 of Miyahara et al are similar to Example 1a. In Example 3a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 30 g of 97% NaOH (0.72 mol NaOH), resulting in an NaOH/NaSH ratio of  $(21.28 + 0.72)/21.28$ , which is equal to 1.033. Example 3b of Miyahara et al is similar to Example 3a. In Example 4a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 50 g of 97% NaOH (1.21 mol), resulting in an NaOH/NaSH ratio of  $(21.28 + 1.21)/21.28$ , which is equal to 1.056. Example 4b of Miyahara et al is similar to Example 4a. Thus, according to the teachings of Miyahara et al, use of an equimolar amount of sodium hydroxide and sodium hydrosulfide in place of the disclosed sodium sulfide results in an NaOH/NaSH ratio above the upper limit of 1.02 required in the process of claim 5.

On the other hand, the present specification shows that when the NaOH/NaSH ratio is above 1.02, the properties of the resulting PAS begin to decline. More specifically, in Example 3 in the specification, wherein the NaOH/NaSH ratio is 1.04 before dehydration (comparable to that of Example 1a of Miyahara et al), and the ratio of NaOH to charged sulfur source is 1.075, the PAS yield is lowered, as is the melt viscosity of the PAS, while the yellow index and the amount of dimer impurity are both undesirably increased. Thus, the modification of the Miyahara et al process to include in situ sodium sulfide formation using equimolar amounts of alkali metal hydroxide and alkali metal hydrosulfide does not meet or render obvious the process requirements of claim 5, and the resulting PAS exhibits a less desirable combination of properties.

Applicants note that the Comparative Examples of Miyahara et al do not employ hydrogen sulfide recycle or additional NaOH. However, Miyahara et al do not provide any teaching or suggestion that would provide one of ordinary skill in the art with any reasonable expectation that a proportion of alkali metal hydroxide to alkali metal hydrosulfide in a ratio of 0.95 to 1.02 mol, in combination with the remaining limitations of claim 5, could be used to result in improved properties in the thus formed PAS.

In this regard, if one of ordinary skill in the art were to following the suggestion of Miyahara et al to form sodium sulfide in situ, as the Examiner has asserted would have been obvious, one of ordinary skill in the art would have naturally considered that at least the following three reactions would occur based on the use of sodium hydrosulfide and sodium hydroxide:





From Miyahara et al, one of ordinary skill in the art would not have known the extent to which each of the above reactions would occur. Further, from Miyahara et al, one of ordinary skill in the art would not have known whether products other than the intended product (sodium sulfide), such as  $\text{H}_2\text{S}$  and  $\text{NaOH}$ , would be contained in the reaction vessel as the result of the step of producing sodium sulfide. In addition, from the Miyahara et al disclosure, one of ordinary skill in the art would not have known what influence these other products would have on subsequent steps, particularly polymerization. One of ordinary skill in the art would normally have feared that these by-products or others in the reaction vessel would adversely affect the PAS product properties. Thus, one of ordinary skill in the art would have considered a step of removing the intended sodium sulfide product from other products obtained in the reaction and dehydration step to isolate sodium sulfide alone for use in the subsequent polymerization.

Presumably, the reason why Miyahara et al did not disclose an example using sodium hydrosulfide and sodium hydroxide “in situ”, despite the knowledge that it would be desirable to use such lower cost reactants, is that various problems have been expected from such a process. This is, in part, described in the present specification as follows:

“There is known a method of using an alkali metal hydrosulfide and an alkali metal hydroxide in combination as materials for the sulfur source. However, this method is difficult to set conditions for stably performing a polymerization reaction. In addition, according to this method, difficulty is encountered on inhibition of side reactions, the content of volatile matter becomes great, and difficulty is encountered on reduction in the content of bis(4-chlorophenyl) sulfide that is an impurity due to a great amount of the alkali metal hydroxide used upon the polymerization reaction” (page 2, lines 15-25).

Thus, even if one of ordinary skill in the art would have considered producing sodium sulfide “in situ” in the dehydration step, anticipated adverse influences on the subsequent

polymerization reaction would have led one of ordinary skill in the art to conduct the in situ formation and dehydration in a different reaction vessel, with separation of the sodium sulfide formed in situ, and Miyahara et al provide no teaching or suggestion that dehydration can be conducted using sodium hydroxide and sodium hydrosulfide as reactants, followed by polymerization in a single reaction vessel as recited in claim 5. In fact, Miyahara et al teach that a system of two or more reaction vessels may be used in order to shorten the cycle time of batch-wise polymerization, thereby teaching away from the present process wherein dehydration can be conducted using the alkali metal hydrosulfide and alkali metal hydroxide, followed by polymerization in a single reaction vessel.

As described in the present specification, the process according to present claim 5 unexpectedly includes formation of a complex of the alkali metal hydrosulfide with the alkali metal alkylaminoalkanoate, providing an unpredictable stability to the reaction which allows dehydration using low cost reactants to be conducted, followed by polymerization in a single reaction vessel:

“In the dehydration step, it is considered that the alkali metal hydroxide reacts with the organic amide solvent by the heat treatment to form an alkali metal alkylaminoalkanoate, and the alkali metal hydrosulfide exists in the system in the form of a complex with the alkali metal alkylaminoalkanoate.” (page 17, lines 6-11).

As a result, the process recited in present claim 5 provides a significant advancement and the polymerization reaction can be stably performed using these low cost reactants, and inconvenient reactions such as thermal decomposition are inhibited (see the specification at page 6, lines 23-26).

With respect to the molar ratio of alkali metal hydroxide to charged sulfur source, the Examiner acknowledged in the Official Action that Miyahara et al's ratio was outside the



claimed range of 1.015 to 1.050, but the Examiner asserted that the value of 1.054 according to Miyahara et al is very close to Applicants' upper limit of 1.050 and therefore a prima facie case of obviousness exists as the claimed range and the prior art ranges do not overlap but are close enough that one skilled in the art would have expected the claimed product and that of the prior art to have the same properties. The Examiner noted that applicant's working examples show similar properties of the final polymers prepared when employing a ratio of 1.020 and 1.050, and the difference between these two ratios having similar properties is 0.030, over seven times greater than the 0.004 difference between the upper limit of the claimed range and the example of Miyahara et al.

However, the Examiner's comments overlook the fact that the molar ratio of alkali metal hydroxide to charged sulfur source is not the only difference between the process of claim 5 and the teachings of Miyahara et al. That is, as noted above, Miyahara et al also fail to disclose the claimed ratio of alkali metal hydroxide to alkali metal hydrosulfide of 0.95 to 1.02 mol, and Miyahara et al fail to disclose that hydrogen sulfide formed during the dehydration is discharged to the exterior of the reaction system. While the Examiner may assert that each difference is minor, these differences in combination with the remaining limitations of claim 5 unexpectedly result in the formation of a complex between the alkali metal alkylaminoalkanoate and the alkali metal hydrosulfide and allow efficient and economic production of a PAS with a desirable combination of properties. Importantly, Miyahara et al provide no teaching, suggestion or recognition of the formation of a such a complex between the alkali metal alkylaminoalkanoate and the alkali metal hydrosulfide or of significant advantages thereof in the production of PAS in a single reactor.

In determining patentability under 35 U.S.C. §103, it is necessary to determine whether there was an apparent reason to combine known elements in the fashion of the claims at issue, *KSR International Co. v. Teleflex, Inc.*, 550 US 398, 418 (2007). Applicants find no evidence of record which would indicate any apparent reason to one of ordinary skill in the art to modify and supplement the teachings of Miyahara et al to result in the process of claim 5. To the contrary, the formation of an alkylaminoalkanoate and the complex of the alkali metal hydrosulfide therewith according to the process of claim 5, and the stability, efficiency and effectiveness in producing high quality PAS of the claimed process in a single reactor are unexpected and surprising in view of the Miyahara et al teachings.

Accordingly, the processes of claim 5, and claims 7, 10-13, 17 and 18 dependent thereon, are nonobvious over Miyahara et al and the rejection under 35 U.S.C. §103 has been overcome. Reconsideration is respectfully requested.

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Please charge any fees required in connection with the present communication, or credit any overpayment, to Deposit Account No. 503915.

Respectfully submitted,

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